Volume 9, Number 7
1009-1963/2000/09(07)/0494-06
CHINESE PHYSICS
July, 2000
© 2000 Chin. Phys. Soc.

STUDY OF THE NUCLEAR PHENOMENA IN THE LOW ENERGY (60–360 keV) PROTON BEAM IMPLANTATION ON METALS*

Wang Tie-shan(王铁山),† Zhu Yong-tai(诸永泰), Wang Zhi-guo(王志国),
Li Song-lin(李松林), Wang Shu-jin(王树金), and Jin Gen-ming (靳根明)
Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, *China*

(Received 20 September 1999; revised manuscript received 21 December 1999)

A series of studies on the nuclear interaction and its effects in solids during proton implantation were performed in the energy region between 60 and 360keV. Some charged particle products were observed in the implantation experiments on metal samples such as deuterated titanium, titanium alloy, titanium foil on molybdenum substrate, molybdenum and stainless steel, etc.. The energies of the charged particle products are about 3.9, 5.6 and 8.4MeV, respectively. These particles were identified as alpha particles with both detector telescope and absorption measurements. These three kinds of alpha products were found from different proton sub-barrier reactions. The exciting curve of this reaction increases exponentially with the growth of proton energy in the energy region from 90 to 330keV. The radiation damages induced by the nuclear products in the sample have been studied with scanning electron microscopy technique. The possible surface damages induced by them were observed evidently. The origin of the nuclear reactions and their effect on implantation are discussed.

Keywords: low energy proton, nuclear reaction, ion beam implantation
PACC: 2540, 1385, 2588, 5240M

I. INTRODUCTION

The interaction between protons and various kinds of matter have been studied for over half a century.[1−9] However, the nuclear reaction induced by protons with energies of only a few hundred keV has not yet been well studied, since very few reactions have been predicted by theoretical analysis.[1,4] Therefore, experimental data in the energy region lower than 500 keV are rare.[5,6] Some laboratories engaged in the study of the energy region for nuclear astrophysical purposes[5−7]. Following the p+Ti^{2}H$_x$ experiment done by OKTAVIAN laboratory, Osaka University, Japan, some charged-particle products were also observed in our 60–330 keV proton implantation experiments.[9] There must be some sub-Coulomb barrier proton reactions occurring in the implantation process. Moreover, three of the reaction products in the p+Ti^{2}H$_x$ experiment exhibit the features of alpha particles. Thus the problem becomes very complex, because the alpha particle cannot be directly produced from the bombarding nucleus (proton) and the major target nucleus (deuteron). These experimental phenomena can always be observed clearly and are always reproducible.[9] Here, a series of experimental studies on these phenomena are reported.

II. EXPERIMENTAL PROCEDURE

Experiments were done with a Cockroft-Walton accelerator. The schematic view of the experimental system is shown in Fig.1.

The proton beam was very pure (purity > 99.9%) after going through a magnetic analyzer. Charged particle spectra were detected by two Si(Au) solid state detectors (SSDs) of about 450 μm in thickness. They were placed at 90° and 135° in the laboratory system to the proton beam direction, respectively. A $\Delta E + E$ telescope was also installed at 90°. All the experimental data were obtained with the detector at 90°, and the measurement at 135° was for checking the angular dependence of the reaction products. The detection solid angles were about 7×10^{-4} sr. The Ti^{2}H$_x$ target was a 2mg/cm^2 deuterated titanium layer on a 0.2 mm molybdenum substrate, which was prepared in China Atomic Energy Institute by special gas loading method. The amount of the loaded deuterium was estimated by the difference of deuterium gas pressure in the chamber before and after the loading process. Hence, the loading ratio of deuterium to titanium was therefore determined by the amounts of deuterium and titanium. The backside of the target was directly cooled by high-pressure water during the experiment, and it could bear over 1 kW ion beam bombarding. The ion beam current was directly mea-

*Project supported in part by the National Natural Science Foundation of China (Grant no. 19845002), the Special Project of the Presidential Foundation of the Chinese Academy of Sciences and also the Special Project of the Nuclear Power Institute of China.
† Corresponding author, E-mail:tswang@ns.lzb.ac.cn

sured from the target, thus the measured value could be higher than the real ion beam current, on considering the secondary electron effect. The charged particle detection system was calibrated with a Thc-Thc' standard alpha source, which contains two kinds of alpha particles with energies 6.15 and 8.78 MeV, respectively. The energy uncertainty in the experiment could be less than 200 keV.

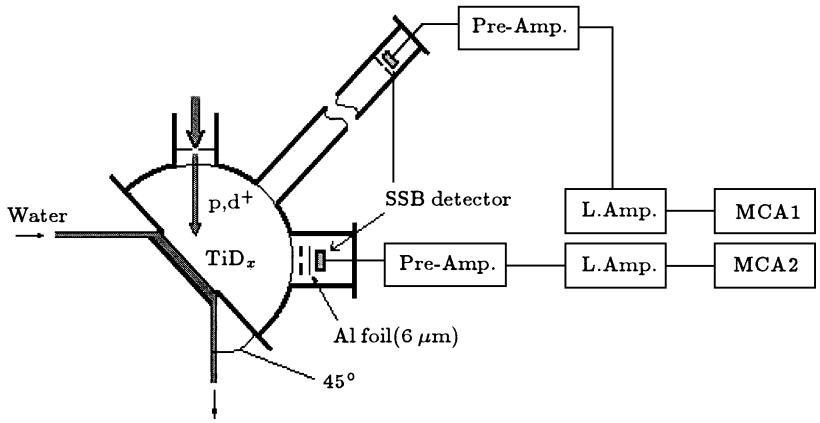


Fig.1. schematic view of the experimental system.

Ⅲ. EXPERIEMNTAL PHENOMENA

A. Charged-particle products

Different kinds of charged particles were observed; their energies can be about 3.0, 3.9, 5.6 and 8.4MeV. A typical charged particle spectrum is shown in Fig. 2.

The peak with about 3.9 MeV had very high intensity. The maximum reaction rate in the experiment reached to over 10^5 r/s at the 324 keV and 1.2 mA proton beam bombardment.

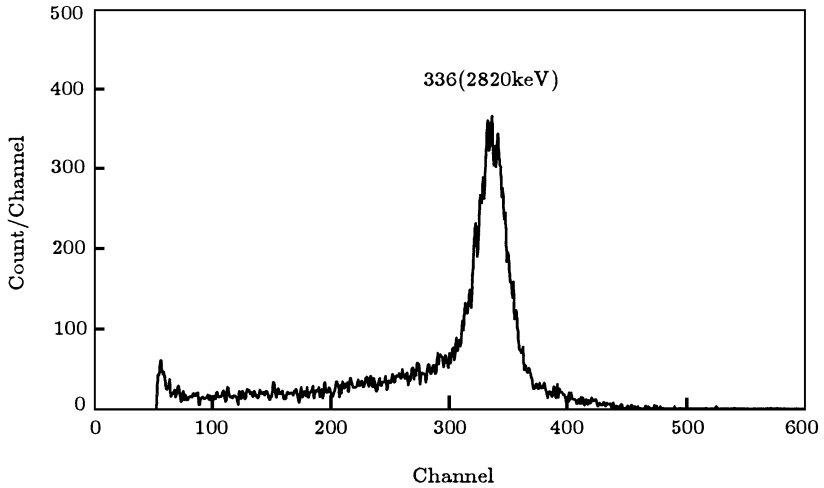


Fig.2. A typical charged particle spectrum in the p+Ti^{2}H$_x$ experiment. Proton energy: 330 keV; beam current: 1.10 mA; measure time: 1000 s; dead time: $\leq$0.2%; Target: Ti^{2}H$_x$. The atomic ratio is $x = {}^2$H/Ti ≥ 1.6; and a 6μm aluminum foil is set in front of the detector.

B. Identification of the charged particle products

The identification experiments were done with both $\Delta E + E$ coincidence and absorption methods. A typical two-dimensional charged particle spectrum is shown in Fig.3.

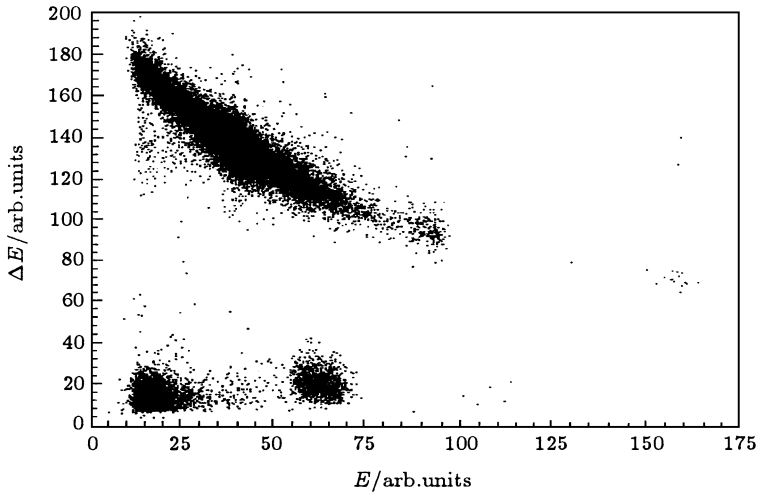


Fig.3. A two-dimensional charged particle spectrum in p + Ti^2H_x experiments.

At least four kinds of charged particles are shown in Fig. 2. One is the 3.0MeV proton from the D(d,p)T secondary reaction induced by the recoiled deuteron. The other three kinds of charged particle products are located in the alpha (or ^{3}He) zone and their energies are 3.9, 5.6 and 8.4 MeV, respectively. It is very difficulty to identify whether the products are ^{4}He or ^{3}He due to the limitation of resolution of the detector telescope system. Therefore, the absorption method is applied to the study of the feature of energy loss in the charged particle products. There is only the shift of peak position but no other difference on the feature of the spectra, when aluminum, nickel and titanium foils with the similar thickness are used. Thus, the unknown peaks are from energetic particles, and the reactions are of true proton beam-target effect, not from an indirect reaction occurring between scattered protons and the screen foil. The dependence of energy loss on aluminum foil thickness is shown in Fig. 4.

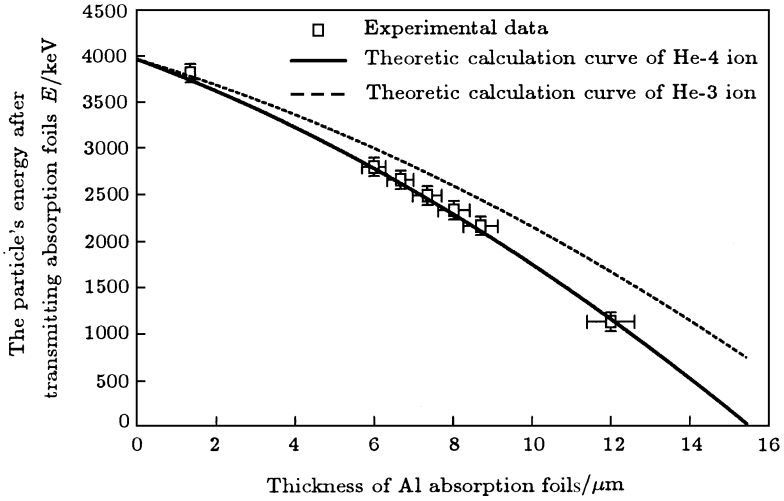


Fig.4. Comparison of the experimental energy loss data measured by changing thickness of aluminum screen foils and the theoretical energy-loss curves of ^{3}He and ^{4}He ions in aluminum.

It is clearly shown in Fig.4 that all the experimental data are located on the theoretical energy-loss curve for alpha particles. Thus, the unknown particle with 3.9 MeV is identified to be an alpha particle but not ^{3}He particle. The original energy of the particle has been easily determined to be 3.9 MeV by extrapolating the energy loss curve.

C. Energy dependence of the reaction rate

Energy dependence of the reaction has been studied in our experiment. The primary excitation curve of the reaction is shown in Fig.5.

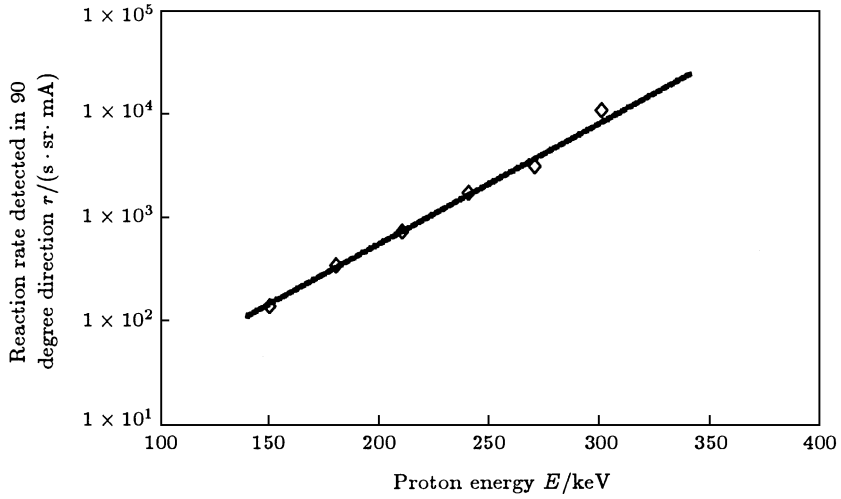


Fig.5. Energy dependence of the detected unknown reaction rate.

The angular distribution was checked by the measurement made at 90° and 135°. The difference of the reaction rates in both directions was difficult to be identified in the uncertainty of the measurement. It was observed that the peak positions did not change significantly while the proton energy was changed from 150 to 330 keV. This implies that the reaction product energies are not evidently energy-dependent in this energy region, since the kinetic energy of the protons was much less than the Q value. Thus, we may estimate roughly the reaction rate with the experimental data measured in the 90° direction. The fitted curve of the experimental data in Fig.6 is linear in semi-logarithmic coordinates. This means that the reaction rate grows exponentially with the increase of proton energy.

D. Observation of the sample surface and impurity analysis

The target surface was observed with scanning electron microscope (SEM). The surface was damaged seriously. A lot of small balls in size less than 1 μm was formed in some areas (See Fig.6).

X-ray photoelectron spectroscopy (XPS) and gas mass spectroscopy (GMS) methods were used to analyze the elements of the target. The contents of the impurity elements are listed in Table 1. The major impurity elements are carbon and oxygen. The ratio of nitrogen is less than 3 percent. Although the elements of silicon, sulfur, sodium, chlorine, calcium etc. are also observed within very tiny ratio.

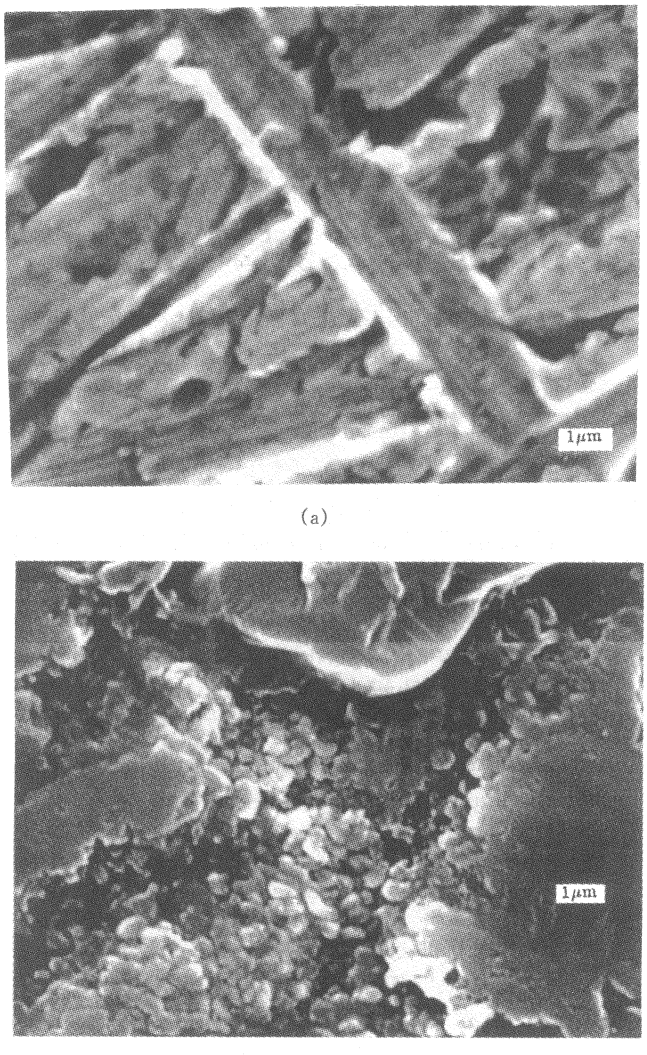


Fig.6. The SEM pictures of the surface of Ti^2H_x target (×10000 times): (a) Surface without implantation; (b) surface implanted with proton.

Table 1. Quantification ratios of the impurity elements measured by XPS.

Peak	Center	Peak area	% (normalized)
C 1s	285.00	1070560	77.36
O 1s	532.58	87756	17.47
N 1s	399.62	9285.8	2.67
Na 1s	1071.98	7862.6	0.895
Si 2p	102.58	910.46	0.351
S 2p	168.84	1751.8	0.392
Ca 2p3	347.94	1591.1	0.178
Cl 2p	199.16	866.70	0.129

E. Other background experiments

Some comparison experiments were done with targets of titanium alloy, titanium on molybdenum substrate, molybdenum and stainless steel etc.. There were only some counts of charged particle products located in the interesting energy range. The reaction rate was at least four orders lower than that in the p+Ti^{2}H$_x$ experiment.

IV.DISCUSSION AND CONCLUSIONS

These reactions cannot be direct reaction between a proton and a deuteron, because the product is alpha particle, whose mass is higher than that of a proton plus a deuteron, therefore, this cannot satisfy the basic principle of energy-mass balance. However, the phenomenon was observed clearly in our experiment. There must be some sub-Coulomb barrier reactions between the proton and nuclei, of which the experimental target material consists other than deuterium.

Another major target constituent is titanium nuclei. All the Q values of the possible reactions between proton and titanium are negative,[1] thus, there is no possibility for these reactions to occur in low energy protons implantation experiments.

The possibility of any reaction occurring between proton and impurity nuclei in the target is therefore considered. The possible impurities in the target are tritium, helium, carbon, nitrogen, oxygen and some other elements. The reactions with energetic alpha product that occur between a proton and the above possible impurity nuclei are:[1,2,7,8]

$$^{15}\mathrm{N} + \mathrm{p} \rightarrow ^{12}\mathrm{C} + \alpha + Q(4.966\mathrm{MeV}), \quad (1)$$

$$^{17}\mathrm{O} + \mathrm{p} \rightarrow ^{14}\mathrm{N} + \alpha + Q(1.191\mathrm{MeV}), \quad (2)$$

$$^{18}\mathrm{O} + \mathrm{p} \rightarrow ^{15}\mathrm{N} + \alpha + Q(3.979\mathrm{MeV}). \quad (3)$$

Among these three reactions, the energies of alpha products from reactions (2) and (3) are 0.926 and 3.14 MeV, respectively; they are much less than the experimental data, hence, these reactions have no effect in the interpretation of experimental results. Reaction (1) seems to be the only candidate, due to the energy of the alpha product being 3.73MeV.[7] However, the natural abundance of ^{15}N is only 0.366%. If its reaction cross section is not above 1 barn, its reaction rate under our experimental condition should be a few orders lower than that of the detected events (once reached 10^5 r/s).[7,9,10] According to the impurity analysis results obtained by XPS and GMS methods, the amount of nitrogen was far from enough to be applied to the interpretation of the experimental phenomena, unless the reaction cross section is larger than a barn, which is impossible according to the theoretical prediction.

Some other nuclear reactions with alpha product between proton and light nuclei, e.g., ^{6}Li(p,α) ^{3}He +4.018MeV, ^{7}Li(p,α) ^{4}He+17.36MeV, ^{9}Be(p,α) ^{6}Li +2.126MeV, ^{10}B(p,α)^{7}Be+1.146MeV, ^{11}B(p, 2α)^{4}He +8.682MeV, ^{11}B(p,α) ^{8}Be+8.59MeV, ^{19}F(p,α) ^{16}O +8.11MeV and ^{27}Al(p,α) ^{24}Mg+1.601MeV are also considered. They are also not the suitable candidate reactions for the 3.9MeV alpha product. The ^{11}B(p,α)^{8}Be+8.59MeV, ^{19}F(p,α) ^{16}O+8.11MeV, ^{7}Li(p,α) ^{4}He+17.36MeV might be used to interpret the high energy alpha particle products, but no significant impurities of ^{11}B, ^{19}F and ^{11}Li were found in the target by the element analysis.

T(d, n)^{4}He reaction has been considered too, because there might be some tritium in the target from the reaction product of D(d, p)T reaction. However, this is already a sub-secondary reaction, and the alpha particle from T(d, n) ^{4}He reaction has only 3.5 MeV energy, lower than the unknown particle's energy, thus it could not be the suitable interpretation. In addition, the experiment done with a fresh target without any tritium displayed the same phenomenon. Thus this possibility is also excluded.

Since there is not any convinced interpretation according to the known nuclear reactions between the proton and possible target nuclei, this reaction is considered anomalous. Some proposals are brought out.

Firstly, D $+^3$He $\rightarrow^4$ He + p + (18.89MeV) secondary reaction is considered, because deuteron and helium-3 produced by the D + p $\rightarrow^3$ He + γ (5.5 MeV) reaction can receive energy by the recoiling of proton and may react each other in the target material. In

the above process, the alpha particle with 3.8 MeV energy and proton with 15.09 MeV should be produced. If this reaction occurs, a peak should appear at around 2.2 MeV, which corresponds with the energy loss of 15.09 MeV proton in a 450μm Si detector. However, we have detected only the alpha particle with about 3.9 MeV without significant peak around 2 MeV. Therefore, this interpretation is also not very convincing.

The neutron reactions, e.g., n+Mo reactions, are also considered. However, the possible neutrons are from D-D secondary reaction, which was observed in less than 10^2 n/s in the experiment. Only a few nuclear reactions can be induced by D-D neutron, and the cross section should be very small. Therefore, the neutron could not play any meaningful role.

Multi-body reaction is also considered, even though the reaction cross section of three-body reaction is considered as over 30 orders lower than two-body reaction based on theoretical prediction.[11] Because the atomic ratio of deuterium to titanium can reach up to 2.0, there is more than one deuterium in each lattice cell of a titanium crystal. When the shell electron of deuterium is captured, the deuterium in titanium exists as deuteron (bare nucleus). Two or more deuterons exist in plasma state with very high density inside lattice (some one called it plasma crystal).[12] In this case, bombarding proton is supposed to react with two or more bounded deuterons under such an experimental condition.[5] One of the proposed reaction is $\mathrm{p} + 2\mathrm{D} \rightarrow {}^4\mathrm{He} + \mathrm{p} + Q(23.8\mathrm{MeV})$. In this process, an alpha particle with 4.7 MeV and a proton with 19 MeV should be produced. The same problem is that no 19.1 MeV proton was detected in our experiment. Also the alpha particle energy from p+2D reaction is higher than the energy detected in this experiment. So, this proposal is also not convincing.

Therefore, the origin of this reaction is still unclear. The $^{15}\mathrm{N}(\mathrm{p},\alpha)^{12}\mathrm{C}$ reaction seems to be the most possible candidate, but the amount of nitrogen in the target is far from enough to be used to support this assumption,[10] unless the cross section of $^{15}\mathrm{N}(\mathrm{p},\alpha)^{12}\mathrm{C}$ is enhanced significantly. Whatever the kind of nuclear reaction is, it must be a reaction under the Coulomb barrier. Because the bombarding nucleus is a proton with very low energy, this reaction might play some role in nuclear astrophysics (solar physics) and the application of fusion energy. The radiation effect should be considered in the implantation experiment. The surface damage of the target is very serious, and a lot of small balls with a size of several hundred nanometers are formed, such large clusters are difficult to be interpreted as the sputtered cluster induced by protons with only hundreds of keV. One possible interpretation is that they are sputtered cluster induced by the nuclear products. At the same time, the ranges of the nuclear product could be much longer than the proton implantation range, thus they could induce some long range effect in the experiment.

ACKNOWLEDGMENTS

We are grateful to Messrs. M. Wan, G. C. Shong and P. Z. Wang, who have operated the accelerator for the experiments. The first author would like to thank Prof. A. Takahashi, Osaka University Japan, for his helpful discussion and information.

REFERENCES

[1] F. E. Cecil, D. Ferg, H. Liu *et al., Nucl. Phys.,* **A539** (1992), 75.

[2] F. K.McGowan, T. Milner, *Atomic Data and Nuclear Data Tables,* **18** (1976),8.

[3] X. T. Lu, Z. C. Chen, J. G. Chen, Handbook of Nuclear Data (Press of Atomic Energy, Beijing, 1981), p.6(in Chinese).

[4] G. Battistig, G. Amsel, E. d'Artemare, *Nucl. Ins. & Meth.,* **B66** (1992), 1.

[5] E. Ray, R.Kirsch, H.H. Mikkelsen *et al. Nucl. Ins. & Meth.,* **B69**(1992),133.

[6] M. Youn. H. T. Chung, J. C. Kim *et al., Nucl. Phys.* **A533** (1991), 321.

[7] C. Rolfs, S. Rondney, *Nucl. Phys.,* **A235**(1974), 450.

[8] H. Lorenz-wirzba, C. Rolfs,W. S. Rodney, *Nucl. Phys.,* **A313**(1979), 346.

[9] T. S. Wang, Y. T. Zhu, Z.G. Wang *et al.,* Proc. of ICACS-18 (Odense, Denmark, Aug. 1999), p.168.

[10] M. Soltani-farshi, H. Baumann, B. Baretzky *et al., MER Syp. Proc.,* **513**(1998), 203.

[11] A. Takahashi, K. Maruta, K. Ochiai *et al., Fusion Technology,* **34**(1998),2734.

[12] Q. Q. Gou, *Chinese J. Atom. and Molec. Phys.,* **15**(1998), 7.